

Insulator to Metal Transition in Shock-Compressed Silica

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Due to its significant abundance in the earth mantle, the behavior of silica, SiO_2 , under high pressure has been extensively studied, both theoretically and experimentally, over the past decades. Up to now, however, these studies have mainly focused on the characterization of the different allotropic phases, α -quartz, cristobalite, and stishovite which are found for the solid state near room temperature (300 K) and pressures generally below 1 Mbar (1 Mbar = 100 GPa). Despite significant progress in our understanding of the behavior of silica in this regime, little is known, however, on its various physical properties for pressures and temperatures above, respectively, 100 GPa and 5000 K.

In the present work, we focus on the description of the electrical and optical properties of silica for pressures ranging from 100 to 1200 GPa and corresponding temperatures which span from 5000 to

90,000 K. This study is motivated by recent measurements of the Al_2O_3 , LiF [1], and SiO_2 [2] reflectivities along the principal Hugoniot using shock compression techniques. These measurements show a significant increase in reflectivity for these three systems, reaching between 20% to 50% at the highest pressures. This smooth rise in reflectivity generally signals a transition from an insulator to a metallic state. At normal conditions, these three crystals present a significant band-gap of about 10 eV. The increase in reflectivity observed experimentally has been modeled for LiF and Al_2O_3 as the combined effect of pressure-induced gap closure and thermal excitation resulting from the increase of both pressure and temperature characteristics of shocked experiments [1]. Despite this attempt, the mechanism leading to the semi-conductor-metal state is still not clear and requires further investigations using *ab-initio* approaches.

To this end, we performed Quantum Molecular Dynamics (QMD) simulations of shock-compressed silica for conditions corresponding to these recent measurements. In QMD simulations, the electrons receive a fully quantum mechanical treatment within the framework of Density Functional Theory (DFT). In contrast to the assumption used for the modeling of the LiF and Al_2O_3 reflectivities [1], we found that the frequency-dependent QMD conductivities do not

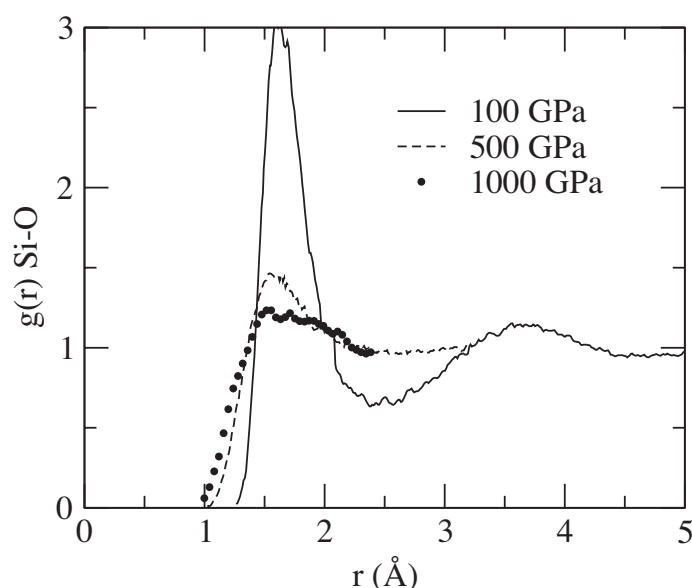


Figure 1—
Reflectivity versus pressure along the principal Hugoniot. Full points are experimental results of P.M. Celliers, et al. [2].

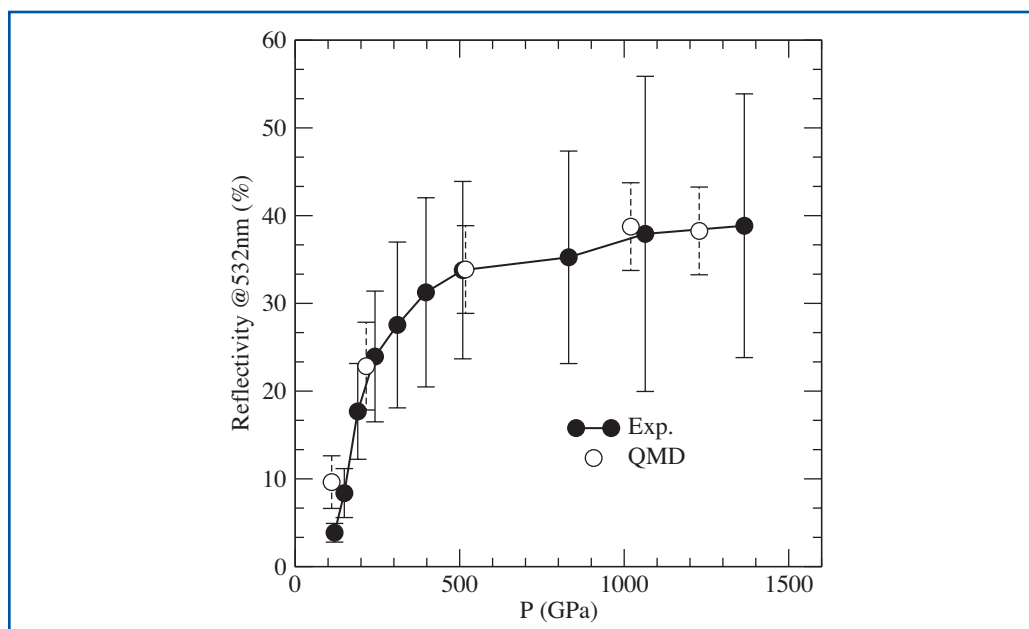


Figure 2—
Pair correlation
function $g(r)_{\text{Si-O}}$ along
the principal Hugoniot.

exhibit a Drude-like behavior. This suggests that a non-ideal metallic state is reached for SiO_2 , even at the highest pressures where measurements were performed. The accuracy of the method at describing the properties of SiO_2 in this regime is corroborated by the remarkable agreement between the QMD and experimental reflectivities above 100 GPa shown in Fig. 1.

To shed some light on the mechanism responsible for the behavior of the reflectivity as a function of increased pressure, we show, in Fig. 2, the variation of the silicon-oxygen pair correlation function, $g_{\text{Si-O}}(r)$, along the principal Hugoniot. At the lowest pressure, $P = 100$ GPa, the pair correlation function, $g_{\text{Si-O}}(r)$, exhibits a sharp maximum at around $1.5a_B$. This shape clearly indicates that the fluid is mostly in a molecular state, with some dynamically broken bonds. At this pressure, the DC conductivity and the associated reflectivity are rather small. As the pressure increases, Fig. 2 shows that the maximum clearly noticeable at 100 GPa has significantly declined at 500 GPa. In the meantime, the DC conductivity and reflectivity sharply increased at this pressure. At 500 GPa and beyond, the silicon-oxygen pair correlation function is representative of an atomic fluid indicating that the SiO_2 molecules have almost completely dissociated. Accordingly, the DC

conductivities and the reflectivity increase only slightly in this pressure range and reach a plateau.

This analysis suggests that a model based on thermally activated processes (TAP) of electrons across a band gap, reduced by the effect of increased pressure, needs to account for the dissociation of the various molecular species present in the media. This direct correlation between the increase in conductivity and the dissociation of molecular systems has already been noticed for other shocked molecular fluids such as hydrogen and nitrogen [3, 4]. The result found here for a more complex system, SiO_2 , indicates that this correlation is rather generic and characteristic of the behavior of shock-compressed molecular fluids [5].

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